

# MASS TRANSFER MECHANISM IN REAL CRYSTALS BY PULSING LASER IRRADIATION

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**ABSTRACT.** The dynamic processes in the surface layers of metals subjected activity of a pulsing laser irradiation, which destroyed not the crystalline structure in details surveyed. The procedure of calculation of a dislocation density generated in bulk of metal during the relaxation processes and at repeated pulse laser action is presented. The results of evaluations coincide with high accuracy with transmission electron microscopy dates. The dislocation-interstitial mechanism of laser-stimulated mass transfer in real crystals is presented on the basis of the ideas of the interaction of structure defects in dynamically deforming medium. The good compliance of theoretical and experimental results approves a defining role of the presented mechanism of mass transfer at pulse laser action on metals. The possible implementation this dislocation-interstitial mechanism of mass transfer in metals to other cases of pulsing influences is justified

## 1. INTRODUCTION

It was appeared experimentally in the works [1]-[3] that the Q-switched laser pulses with power density  $q_i = 10^6 \div 10^7 W/cm^2$ , pulse duration of  $\tau = 10 \div 50 ns$  stimulate anomalous deep atom penetration from irradiated metal surface to the depth value of which is considerably more than thermal influence depth. To interpret this phenomenon of high atomic mobility, a mechanism of interstitial atom migration under the conditions of high-rate strain was presented in [1]. This mechanism cannot interpret observed experimentally long-range mass transfer, because the interstitial atoms are short-live defects, which quickly eliminated on sinks. To escape this problem the authors of the work [4] supposed interstitial atom dragging by the dislocations, which were moving by the thermal stresses. In this case lifetime of the complex "*dislocation+interstitial atom*" was sufficient for right estimations of the penetration depth. Thermal stresses were calculated using the exact solution [5] of the thermal conductivity problem in the laser-influenced metal. The calculations show a possibility of generation of the dislocations during a relaxation of the deformations in the metal. The deformation processes during the laser actions were confirmed by the presence of topographical curving structure and formation of polygonal grids in crystalline *Mo* [6], and cellular dislocation structures in *Armco-Fe* [7]. Dislocation structure analysis, which was performed with the data presented in the work [8], has evidenced the possibility of generating large amount of point defects of interstitial type under these conditions. Model calculations, which

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were performed in the work [4], show the dynamical state of the laser-influenced metal. This state is stipulated by the moving of the gradients of thermal rapid-changed stresses from the skin layer into the bulk of the metal. These stresses cause the compressing waves, even under special conditions shock wave forming, and penetration in considerable depth of laser-influenced metals. The formation of polygonal grids in *Mo* [6], and cellular dislocation structures in *Armco - Fe* [7] demonstrate the interaction between dynamic stresses and dislocations, which are transferred in dynamically deformed lattice.

In the present work we discuss the problem of the creation, annihilation, and lifetime of the "*dislocation+interstitial atom*" complex in the frame of the dislocation-interstitial mechanism in the metals, which are influenced by pulsing laser irradiation. The results permit correct interpretation of anomalous deep atom penetration from the laser-affected surface to the bulk of the metal on distances of tens of micrometers.

## 2. BASIC RELATIONS

Pulsing laser heating of the near-to-surface layers of metals stipulates the rapid thermal expansion of these layers. Heat velocity is connected with the velocity of the temperature change with the time  $t$  :

$$\partial_t T \approx 10^6 \div 10^{10} K/s$$

which, in turn, depends on the duration of the laser pulse. It was shown in the work [9], that in the case of duration of laser pulse  $\tau \approx 10^{-10} s$  the stress fields in the influenced metal one can describe to solve the dynamic thermal-elasticity equations [10] in the quasi-stationary approach [11].

The temperature  $T(r, \phi, z, t)$  in bulk of the metal ( $z > 0$ ), which is irradiated by the laser beam with the width  $\beta$ , intensity  $I_0$ , duration  $\tau$ , and skin layer  $\kappa$ , is described by the equation [5]:

$$\partial_t T - a\Delta T = I_0 e^{-\beta r^2 - \kappa z} \begin{cases} 1, & \text{if } t \leq \tau, \\ 0, & \text{if } t > \tau, \end{cases}$$

with boundary conditions:

$$(\partial_z T)_{z=0} = hT(r, 0, t).$$

Here  $a$  is thermal conductivity coefficient;  $I_0$  is defined as

$$\frac{q_0}{k} a \eta,$$

where  $q_0$  is the power of irradiation,  $\eta$  is the absorption coefficient,  $k$  is thermal capacity coefficient;  $h$  describes the heat flow absorbing;  $\Delta$  is the Laplasian. We omit below coordinate  $\phi$  due to the cylindrical symmetry of the considered problem.

Exact solution of this problem is:

$$(1) \quad \begin{aligned} T(r, z, t) = & \frac{I_0}{2\beta} \int_0^\infty \xi J_0(\xi r) e^{-\frac{\xi^2}{4\beta}} \{ F(\xi, t) - \\ & - \begin{cases} 0, & \text{if } t \leq \tau \\ F(\xi, t - \tau), & \text{if } t > \tau \end{cases} \} d\xi. \end{aligned}$$

where

$$\begin{aligned}
F(\xi, t) = & \frac{1}{a(\kappa^2 - \xi^2)} \{ e^{-\kappa z} (e^{a(\kappa^2 - \xi^2)t} - 1) + \\
& + (h + \kappa) \left[ \frac{e^{-\xi z}}{2(h + \xi)} \operatorname{erfc} \left( \frac{z}{2\sqrt{at}} - \xi\sqrt{at} \right) \right. \\
& - \frac{e^{\xi z}}{2(\xi - h)} \operatorname{erfc} \left( \frac{z}{2\sqrt{at}} + \xi\sqrt{at} \right) - \\
& - \frac{1}{2} e^{a(\kappa^2 - \xi^2)t} \left( \frac{e^{-\kappa z}}{\kappa + h} \operatorname{erfc} \left( \frac{z}{2\sqrt{at}} - \kappa\sqrt{at} \right) \right. \\
& - \left. \left. \frac{e^{\kappa z}}{\kappa - h} \operatorname{erfc} \left( \frac{z}{2\sqrt{at}} + \kappa\sqrt{at} \right) \right) \right] + \\
& + \frac{h(\kappa^2 - \xi^2)}{(\xi^2 - h^2)(\kappa^2 - h^2)} e^{zh + a(h^2 - \xi^2)t} \times \\
(2) \quad & \times \left. \operatorname{erfc} \left( \frac{z}{2\sqrt{at}} + h\sqrt{at} \right) \right] \}.
\end{aligned}$$

The solution (1) of this problem reveal the maximum of the axis component of thermal-elastic stresses  $\sigma_{zz}(0, z, t)$  [12]. This maximum is formed at the depth of the metal to the end of laser pulse. The value of the maximum, and the value  $z_0$  depends on the parameters of laser irradiation, and the physical properties of irradiated metal. The value of the maximum reach the extremum and it position approach to the surface when the exposure time is decreased [12]. On the surface  $z = 0$  axis component  $\sigma_{zz}(0, 0, t) = 0$ . It appears because replacement of the surface in  $z$ -up-direction results in a relaxation of axis stresses. In-plane replacements cause at the irradiated surface  $z = 0$  radial-symmetric stresses  $\sigma_{rr}, \sigma_{\phi\phi}$  [9], which are maximal at the surface [13] and depend on the depth  $z$  as [9]:

$$\sigma_{rr} = -\frac{E}{1 - \nu} \alpha \nabla T(r, z, t),$$

where  $\alpha$  is the thermal expansion coefficient;  $E$  is the Young's module;  $\nu$  is the Poisson's coefficient.

As it was shown in [12], stresses, which appeared in laser-irradiated metal, cause a deformation of a lattice. A process of the creation of the dislocations will begin in the volume, which deformation value is more than critical one, exactly, the yield stress. These areas of deformed bulk of a metal are determined by the temperature inhomogeneity Eq.(1) [5]. The processes of the creation of the dislocations are developed due to an action of the different nature sources, usually in combination.

Stretched area will appear near the center ( $r = 0$ ) of the laser spot on the surface ( $z = 0$ ) of the irradiated metal, and the compressed area will form at some distance from the center of laser spot. Calculated stresses [12], which correspond this influence, are shown on the Fig.1.

New generated, and existent in a bulk of the metal, dislocations are influenced by the inhomogeneous thermal-elastic stresses. As a result, effective forces, which appears due to the action of the inhomogeneous thermal-elastic stresses, cause the displacements of the part of dislocation or whole piled up dislocation group, which, in turn cause the plastic deformation of the near-to-surface areas of the laser-influenced metal. The transmission electronic microscopy data show [7], that the formation of the cellular structure is the result of the creation, accumulation,

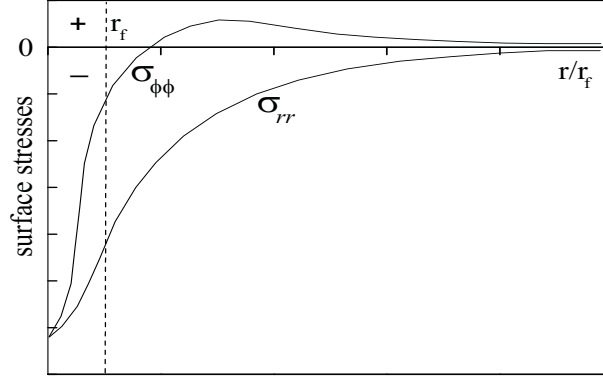


FIGURE 1. Stresses on the surface of a metal, irradiated by a pulsing laser beam with the spot radius  $r_f$

redistribution, of the dislocations, on which the number of the kinks is considerably increased. This formation depends on the number of the laser shots. Observed experimentally increasing of the dislocation loops is connected with the generation of the point defects under described influences.

A motion of the dislocations and its interactions also cause the generation of point defects [4]. A type of generated defect, namely, a vacancy or an interstitial atom, depends on as the dislocation interaction mechanism [14], [15], as the deformation velocity [16]. Generated vacancies and interstitial atoms are considerably non-equilibrium defects. These defects tend to annihilate and disappear in different sorts of the sinks, such, for example, as the dislocations, a surface, grain boundaries [17].

### 3. CALCULATIONS AND ESTIMATIONS

As it was admitted above, appeared in pulse laser influenced near-to-surface layers of a metal thermal gradient cause the thermal stresses. A relaxation of these stresses is realized by the generation of moving dislocations during the time when the value of these stresses is more than value of a yield stress [5]. Therefore one has taken into account an increasing of the number of dislocations during as the thermal stresses relaxation time, as a many-fold action. The analysis in [12] shows, that the deformation due to the pulse laser irradiation of a metal is local deformation. A half-width of deformation curve is greater than a half-width of the laser beam in the case of large values of  $\tau$ , and difference between these half-widths tends to zero when  $\tau$  is decreased. It is clear, that the defect distribution will correlate with these tendencies. As it follows from the results of the works [5], [9],  $z$ -axis component of the density of the dislocations in a metal after laser irradiation is:

$$(3) \quad \rho_{i+1} = \rho_i - \frac{2\alpha}{(1-\nu)b} \int_1^n \nabla T(0, z, \lambda t) d\lambda,$$

where  $i$  is the number of laser shots ( $i = 0, 1, \dots$ );  $b$  is the value of Bürger's vector of a dislocation;  $\rho_0$  is the initial density of dislocations; by the factor  $n$  we take into account the processes, which delay a creation of dislocations by thermal stresses. Calculation of the dislocation density Eq.(3) was carried out for the case of *Armco-Fe*

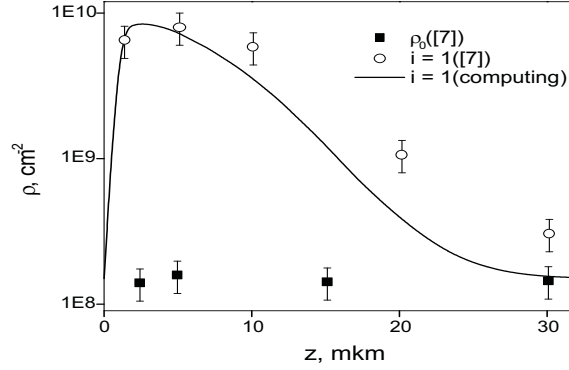


FIGURE 2. Dislocation density in laser irradiated *Armco-Fe*. Experimental points are obtained at Q-switched laser pulse duration  $\tau = 5 \times 10^{-8}s$ .

influenced by Q-switched laser pulse with  $q_0 = 10^7 W/cm^2$ ;  $\tau = 50ns$ ;  $r_f = 0.2cm$ . Results of these calculations at  $h = 0$  are shown on the Fig.2 (continuous line). Experimental data are shown on the Fig.2 also. Experimental points were obtained in [7] by a measuring of the density of dislocations by the method of electron microscopy in *Armco-Fe* sample, which was laser irradiated. Calculations were made in supposition of gathering of the dislocations during relaxation processes. Best correlation of the calculation and experimental data was achieved at the relaxation time of order  $1.6\mu s$ . Therefore we took this time as a time of dissipation of the energy, which was pumped by a laser irradiation into the metal. During this time a generation and motion of the dislocations are ended. Small discrepancy between exact solution Eq.(1) and real experimental data one can see in a decreasing part of the curve Fig.2. This discrepancy occurs due to the volume stresses  $\sigma_{rr}$  and  $\sigma_{\phi\phi}$ , values of which may be large enough to create additional notable append to the dislocation density. We have taken into account also a result of non-linear depth dependence of a dislocation creation process, which is facilitated in the less-defective bulk metal region. Interstitial atoms and vacancies are created during a plastic deformation of the metal. The difference of its creation energies in a deformed crystal is not determining factor for concentration ratio of these point defects [18]. You have taken into account a dynamics of a dislocation interaction, i.e. it velocity  $V$ , stresses  $\sigma$  parameters (deformation value  $\varepsilon$ , velocity  $\dot{\varepsilon}$ .) Moving skew dislocations create interstitial atoms. The velocities of these dislocations are greater than critical velocity when the stresses exceed its critical value [19]. It appears on the early stages of crystal deforming, when the density of the dislocations is low, and number of the steps, which create interstitial atoms, exceeds not the number of the steps, which create vacancies [16]. Point defects in crystals are less movable than the dislocations [19]. Transfer of the dislocations under influence of the internal or external forces may results in mass transfer. Authors of the work [20] supposed, that this mass transfer is a result of interaction of moving dislocations and interstitial atoms. Temperature dependence of a preference parameter for dislocations was calculated in the work [21]. It was shown, that in the temperature interval  $400 - 900K$  interstitial atoms appear in the dislocation core, and vacancies are in its atmosphere. It is clear, that in the case of the edge dislocation, the space

under the dislocation line is preferable for interstitial atom position. When this dislocation will move in the stresses field along its glide line, the interstitial atom will move with dislocation in the place of energy minimum. This atom transfer will take place until the realization of another, more energetically preferable situation will be realized.

First, atom annihilates on the sink. This case is trivial and we will not consider it here.

Second, in the case, when the value of the stresses exceeds the value of Pierl's stress, the non-activated motion of the dislocations is possible. The velocities of this motion may be near the sound velocity [22]. At the some critical velocity value  $V_c$  dislocation may loose the atom. Let us estimate this velocity. We suppose, that a complex "*dislocation + interstitial atom*" will exist until its kinetic energy is less than the link energy of the interstitial atom with the dislocation. We can write:

$$(4) \quad \frac{1}{2}MV_c^2 \geq W,$$

where  $M = m^* + m$ ,  $m$  is the atom mass;  $m^*$  is the dislocation effective mass, which is determined as [19]:

$$(5) \quad m^* = m \frac{2d}{l},$$

where  $d$  is the distance between glide planes;  $l$  is the half-width of a dislocation:

$$l = \frac{d(3 - 2\nu)}{4(1 - \nu)}.$$

Hence we get:

$$(6) \quad V_c = \sqrt{\frac{2W(3 - 2\nu)}{m(11 - 10\nu)}}.$$

In the case of the *Armco - Fe*:  $W = 0.2 \div 0.5 \text{ eV}$  [23];  $\nu = 0.28$ ;  $m = 1.66 \times 10^{-24} M_{Fe} = 9.27 \times 10^{-23} g$ , and

$$V_c = (450 \div 716) \times 10^2 \text{ cm/s}$$

Now it is easy to see, that in the case of free motion with the velocity  $V_c$  during the time  $t_M = 1.6 \times 10^{-6} s$  (relaxation time) complex "*dislocation + interstitial atom*" will be on the distance of  $720 \div 1146 \mu m$ . Experimentally measured depth of the atom penetration from initial layer of  $1 \mu m$  is in order of  $10 \mu m$  [1], [2] (at Q-switched laser pulse duration  $\tau = 3 \times 10^{-8} s$ ), which is considerably less than the above mentioned distance. This difference is caused by an interaction of the dislocations with different defects during a motion through the real crystal. A dissipation of the complex energy results in a decreasing of the velocity of the complex. The defects, which cause energy dissipation in real crystals, are other dislocations, point defects, disperse constituents of other phase, etc. [19]. To estimate these losses let us consider the motion of the complex "*dislocation + interstitial atom*", which mass is  $M$ , under the influence of the stresses forces  $F = f(\sigma)$ , in the media with the damping coefficient  $\gamma$ .

During a laser pulse ( $t \leq \tau$ ) motion equation

$$(7) \quad d_t V_1 = \frac{F}{M} - \frac{\gamma}{M} V_1$$

solution is:

$$(8) \quad V_1 = \frac{F}{\gamma}(1 - \exp\{-\frac{\gamma t}{M}\}),$$

which presents maximum velocity

$$(9) \quad V_{max} = \frac{F}{\gamma}(1 - \exp\{-\frac{\gamma \tau}{M}\}).$$

When the stresses are not influence the motion under consideration, in other words, when the laser pulse is ended, a complex "*dislocation + interstitial atom*" reaches the maximal value of the velocity, which is described by the equation

$$(10) \quad d_t V_2 = -\frac{\gamma}{M} V_2,$$

which solution is

$$(11) \quad V_2 = V_{max} \exp\{\frac{\gamma}{M}(\tau - t)\}.$$

Averaging of the given solutions over the relaxation time  $t_M$

$$< V > = \frac{1}{t_M} \int V(t) dt$$

gives

$$(12) \quad < V > = \frac{F}{\gamma t_M} \left\{ \tau + \frac{M}{\gamma} [1 - \exp\{\frac{\gamma \tau}{M}\}] \exp\{-\frac{\gamma t_M}{M}\} \right\}$$

In the case of large values of  $\gamma$

$$< V > \approx \frac{F \tau}{\gamma t_M},$$

and a ratio

$$(13) \quad \frac{< V >}{V_{max}^{(0)}} \approx \frac{\tau}{t_M} \approx 0.02.$$

Here

$$V_{max}^{(0)} \equiv \frac{F}{\gamma}.$$

Taking into account given ratio, and putting  $V_{max}^{(0)} = V_c$ , we can get for a real depth of surface atoms mass transfer the value

$$z = < V > t_M \approx V_c \tau \sim 14 \div 22 \mu m.$$

"*Dislocation + interstitial*" complex will accelerate to the velocity of the order  $V_c$  at the initial period of a laser influence, i.e. at the times  $\tau/2$  (if the Q-switched laser pulse form is near to triangle). It gives complex decomposition depth  $\sim 7 \div 11 \mu m$ , which is in a good agreement with the experimental data. Better correlation is achieved by taking into account the possibility of the annihilation of the dislocations. The dislocation annihilation takes place when the opposite sign dislocations collide in the process of the motion in parallel glide planes [15]. Simplest annihilation realized for the dislocations with a free core. In the case of the "*dislocation+interstitial atom*" complex annihilation is low-possible because interstitial atoms in the core of dislocation enable this effect. For an edge dislocation another velocity decreasing factors may be the delay of the kinks on the steps of a dislocation, and on the vacancies, which are situated on the edge of the extra plane.

We have admit, that the tagged atoms (radioactive isotopes of  $Fe$ ) are concentrated in smooth near-to surface layer of experimentally investigated samples. Therefore the ratio of penetration depth of tagged atoms to whole depth of high dislocation concentration is proportional to initial ratio of value of tagged atoms layer and value of influenced thickness of a sample.

We can estimate the depth of vacancy atmosphere loosing by a dislocation, using the Eqs. (6), and (13). These events will be realized on the depth  $\sim 3\mu m$  if we put the "*vacancy+edge dislocation*" binding energy value equal to  $0.04eV$  [23]. It is clear, that binding energy "*dislocation + interstitial atom*" exceeds the binding energy "*dislocation + vacancy*", therefore during the motion of complexes with the velocities, values of which are more than the critical value, the dislocations loose first of all the vacancy atmosphere and then the interstitial atoms, which are bounded to the core of the dislocations. This difference in the behavior of the vacancies and interstitials is thermal activated and realizes in the considered temperature interval. Calculated in [5] temperatures in the metal, which was influenced by pulsing laser irradiation, are in good agreement with this idea. Calculations in [5] and experimental data in [24] show that temperature influence of the laser beam during nano-second reaches the depths in order of  $\mu m$ . As a result the concentration of the vacancies in the near-to-surface layers are much than the concentration of the interstitials and vise versa in deepest layers [4] .

#### 4. DISCUSSION

Non monotonous dependencies of the distribution of tagged atoms under surface of pulse laser irradiated metal are in good agreement with the calculations, which were carried out on the base of a concept of the presented above mechanism of capturing and transport of the point defects by the dislocations, which are generated and moving by the thermal fields, which appeared in surface layer of irradiated metal.

The kinetic of a gathering of the tagged atoms  $^{55}Fe + ^{59}Fe$ , which were penetrated from the surface to bulk of a metal, which was influenced by many-fold pulse laser irradiation ( $\tau \sim 10^{-8}s$ ,  $W \approx 0.02J$ ), was investigated in *Armco-Fe* [25], [29]. A maximum of the concentration of radioactive atoms  $^{55}Fe + ^{59}Fe$  began to be formed after the first three cycles of an irradiation. It correlates with  $X$ -ray and electron microscopy data, which shows maximal increase of defects in a metal after the first cycles of pulse laser influence. A displacement of the defect concentration maximum to the depth of the irradiated sample was observed simultaneously with the forming of the maximum. These processes saturation are observed at the number of laser shots  $\sim 10$ . At this time an increasing of the dislocation density in *Armco-Fe* was ended, and new dislocations were gathered into cellular structure, as it was shown by the electron microscopy experimental data [7] .

Effective diffusion coefficients  $D_i^*$  were obtained from the analysis of the tagged atoms concentration experimental data, which were measured in the influenced by pulsing many-fold laser irradiation areas of *Armco-Fe*. The value of  $D_i^*$  is monotony decreased with the number of laser shot  $i$  [24]. This effect is caused by an increasing of the number of the defects, which are created by laser influence. It results in the effective slowing of the motion of the complexes *dislocation + interstitial atom*. A value of  $D_i^*$  is decreased not so fast after  $5 \div 8$  shots. This

slowing is connected with a beginning of the process of effective redistribution of the radioactive isotope [7].

We can describe the experimental data of the works [1], [2] from the positions of the presented above *dislocation + interstitial* mechanism taking into account the data of the work [26]. Compression wave appears in the metal during the time of influence of pulsing laser irradiation. Stresses wave slope is increased during this laser influence on a metal. A number of defects of crystalline lattice is increased due to appearance of the inhomogeneous stresses as a result of laser influence. New defects may be the sinks for migrating from the surface interstitials. Tagged atoms concentration is maximal on the depth  $z_0 \geq \sqrt{a\tau}$ , which coincides with the depth of maximum of thermal stresses at the moment of the end of laser pulse. The number of sinks is decreased with the depth because the distance to the source of the stresses increased, and amplitude of the stresses is decreased. Each next laser pulse pumps radioactive isotopes from the near-to-surface layers to the bulk of the metal, and increases the number of sinks for interstitials. It is reflected on the slopes decreasing of the exponential parts of tagged atom concentration curves as a function of  $i$  [2]. Hence a decrease of the value of  $D^*$  is slowing.

Presented above phenomenological description of the laser-stimulated *dislocation + interstitial* mass transfer process in *Armco - Fe* is confirmed by the experimental data of the investigation of self atom transport in *Ni* [27]. It appeared, that increasing of the vacancy number slowing mass transfer. This phenomenon is not possible to explain in the frame of the vacancy mechanism because an increasing of the number of the vacancies facilitates a diffusion of the atoms in a crystalline lattice. Observed phenomenon is explained easy by the *dislocation + interstitial* mechanism of a pulse influenced mass transfer. At the beginning of a mass transfer migrating interstitials are high movable and interact not practically with the vacancies, which are traps for its. Loosing of a kinetic energy by the interstitials makes its more suitable for vacancy trapping, by the other words a recombination probability increase. This recombination reflects a mass transfer last stadium, which is characterized by exponential dropping parts of the concentration curves.

Presented above estimations and analysis show the nature of pulsing laser stimulated mass transfer in metals. This mass transfer is carried out by the near-to surface layers interstitials, which interact with the moving dislocations during the laser influence and relaxation of stressed state of a crystalline lattice. Taking into account that the stressed state relaxation processes are common for the all pulsing influences [28], which not destroy the structure of an influenced metal, we can state an universality of the presented mass transfer mechanism.

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MASS TRANSFER MECHANISM IN REAL CRYSTALS BY PULSING LASER IRRADIATION1

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